

This listing of claims will replace all prior versions and listings of claims in the application:

**LISTING OF CLAIMS**

-1- (Currently Amended)

A hybrid mesoporous silica composition comprising a framework structure defining the mesopores which is in one domain lamellar or hexagonal and in another domain with wormhole pores and wherein the domains are intergrown together, wherein the silica is defined in anhydrous form, the silica has the formula:



wherein  $1.0 \geq w \geq 0$  and  $1.5 \geq x \geq 0$  and wherein M when present is one or more metal ions.

-2- (Currently Amended)

A composition which is a hybrid wormhole and lamellar or hexagonal framework molecular sieve silica prepared by a neutralizing reaction in an aqueous solution of amine surfactant; a reactive silica species of pH between 5.0 and 10.5; aging of the solution to precipitate the silica and removing of the silica from the solution, wherein the silica is defined in anhydrous form, the silica has the formula:



wherein  $1.0 \geq w \geq 0$  and  $1.5 \geq x \geq 0$  and wherein M when present is one or more metal ions.

-3-(Original)

A composition which is a hybrid wormhole and lamellar or hexagonal molecular sieve silica prepared by a process which comprises:

5 (a) acidifying an aqueous solution of an amine surfactant as a structure director with an acid selected from the group consisting of organic, mineral and oxy acids;

10 (b) preparing a reactive silica species in the aqueous solution by neutralization of a basic soluble silicate solution by mixing with the acidified amine surfactant aqueous solution of step (a) reaching a final pH of about 5 to 10.5;

15 (c) aging the reactive silica species from step (b) at a temperature greater than -20°C;

(d) recovering a solid product from the aqueous solution by removal of the solution; and

20 (e) removing the surfactant from the solid by calcination at 600°C in air for not less than 30 minutes, by solvent extraction, or by treatment with a stoichiometric amount of aqueous acid solution and washing with water, to produce the molecular sieve silica, wherein silica possesses framework-confined mesopores with pore diameters ranging from 1.0 to 12.0  
25 nm, the framework-confined channel structure comprises a hybrid wormhole and lamellar or hexagonal framework

morphology has at least one resolved powder x-ray reflection corresponding to a pore-pore correlation spacing of 1.5 to 15.0 nm, inorganic oxide wall thickness of greater than 0.5 nm, specific surface areas of 400 to  
5 1400 m<sup>2</sup>/g and framework pore volumes of 0.1 to 3 cc/g N<sub>2</sub>.

-4-(Original)

The composition of Claim 3 wherein the silica in step (b) is sodium silicate "water glass" with a SiO<sub>2</sub>/Na<sub>2</sub>O = 1.5 to 4.0.

-5-(Original)

The composition of Claim 3 wherein silica in step (b) is colloidal silica or fumed silica.

-6-(Original)

The composition of Claim 5 wherein soluble silica solution is prepared with addition of an alkali, or organic base to dissolve silica at a high pH greater than 12.

-7-(Original)

The composition of Claim 3 wherein said acid is selected from the group consisting of:

HX where X = Cl, Br, I;

5 H<sub>x</sub>Y where Y = NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>-2</sup>, PO<sub>4</sub><sup>-3</sup>, CO<sub>3</sub><sup>-2</sup> and x equals the charge on Y; and

HZ, where Z = an organic carboxylate, phenolate, citrate, glycolate.

Claim 8 (Cancelled)

-9-(Original)

The composition of Claim 8 having at least one resolved X-ray reflection and an X-ray diffraction pattern selected from the group consisting of Figures 1, 4 and 5.

-10-(Original)

The composition of Claim 8 having a N<sub>2</sub> adsorption-desorption isotherm selected from the group consisting of Figures 2 and 6.

-11-(Original)

The composition of Claim 8 having a BET surface area between 400 and 1400 m<sup>2</sup>/g.

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Amdt. Dated: November 13, 2003  
Reply to Office Action of 10/07/2003

-12-(Original)

The composition of Claim 8 having a textural mesopore volume from 0.01 to 3 cc/g.

-13-(Original)

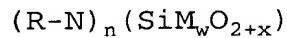
The composition of Claim 8 having TEM micrograph selected from the group consisting of Figures 3A, 7A and 8A.

-14-(Original)

The composition of Claim 8 wherein the silica contains a hexagonal framework structure.

-15-(Original)

The composition of Claim 1 wherein said oxide has a composition as follows:



5 wherein  $(SiM_wO_{2+x})$  is written in anhydrous form without water, wherein R-N is at least one of a selection of neutral aliphatic amines or polyamine surfactants wherein when R-N is present, n is between about 0.05 and 2; wherein when M is present at least one element selected from the group comprising B, Ge, Sb, Zr, W, P, Ba, Y, La,  
10 Ce, Sn, Ti, Cr, Nb, Fe, V, Ga, Al, Zn, Co, Ni, Mo and Cu and w and 2+x are the molar stoichiometries of M and "O", respectively, wherein w is 0.00 to 0.30; x is 0.00 to 1.50.

-16-(Original)

The composition of Claim 15 having a X-ray diffraction pattern selected from the group consisting of Figure 4 wherein the main diffraction peak corresponds to a basal spacing between 2.0 and 15 nm.

-17-(Original)

The composition of Claim 15 in which the surfactant has been removed from the silica matrix by calcination in air at 600°C.

-18-(Original)

The composition of Claim 17 having a N<sub>2</sub> adsorption-desorption isotherm, the shape of which is as in Figure 2.

-19-(Original)

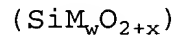
The composition of Claim 15 having a TEM micrograph selected from the group consisting of Figures 3A, 7A and 8A.

-20-(Original)

The composition of Claim 15 in which the surfactant has been removed from silica by solvent extraction or by extraction with an acid.

-21-(Original)

The composition of Claim 1 wherein said silica has a composition as follows:



wherein  $(\text{SiM}_w\text{O}_{2+x})$  is written in anhydrous form without water, wherein when M is present at least one element selected from the group comprising B, Ge, Sb, Zr, W, P, Ba, Y, La, Ce, Sn, Ti, Cr, Nb, Fe, V, Ga, Al, Zn, Co, Ni, Mo and Cu and w and 2+x are the molar stoichiometries of M and "O", respectively, wherein w is 0.00 to 0.30; x is 0.00 to 1.50.

-22-(Currently Amended)

The composition of Claim 1 ~~21~~ having a X-ray diffraction pattern as in Figure 1 or Figure 5.



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-23- (Currently Amended)

The composition of Claim 1 ~~21~~ in which the  
surfactant has been removed from the silica by  
calcination in air.

-24-(Original)

The composition of Claim 23 having a N<sub>2</sub> adsorption-desorption isotherm shape as in Figure 2 or Figure 6.

-25-(Original)

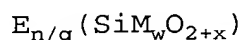
The composition of Claim 23 having a TEM micrograph image selected from the group consisting of Figures 3A, 7A and 8A.

-26-(Original)

The composition of Claim 23 in which the surfactant has been removed from the silica matrix by solvent extraction or extraction with an acid.

-27-(Currently Amended)

The composition of Claim 1 wherein said silica has a composition expressed in anhydrous form as follows:



5 where E is one or more exchange ions, q is the weighted molar average valence of E; n/q is moles of E per mole of Si, n is the charge on the composition excluding E, and w and 2+x, respectively, are the molar compositions of M and oxygen in the framework, wherein  $1.0 \geq w \geq 0$  and  $1.5 \geq x \geq 0$ .

-28-(Original)

The composition of Claim 27 having a X-ray diffraction pattern selected from the group consisting of Figures 1 and 5.

-29-(Original)

The composition of Claim 27 in which the surfactant has been removed from the silica by calcinations in air.

-30-(Original)

The composition of Claim 29 having a N<sub>2</sub> adsorption-desorption isotherm shape as in Figure 6.

-31-(Original)

The composition of Claim 29 having a TEM micrograph selected from the group consisting of Figures 3A, 7A and 8A.

-32-(Original)

The composition of Claim 27 in which the surfactant has been removed from the silica by solvent extraction or by extraction with acid.

-33-(Original)

The composition of Claim 27 having a N<sub>2</sub> adsorption-desorption isotherm shape selected from the group consisting of Figure 6.

-34-(Original)

5 The composition of Claim 1 having a TEM micrograph of Figure 8A showing ordered pore structures in a hexagonal unit cell within small particle materials and having a selected area electron diffraction pattern showing polycrystalline ordering in the silica as seen by multiple diffraction spots as shown in Figure 8C.

-35-(Original)

A composition which is a hybrid molecular sieve silica prepared by a process that comprises:

(a) preparing an aqueous solution of a amine surfactant as an organic structure director;

5 (b) adding a basic soluble silicate to the amine solution;

(c) neutralizing the basic amine and silicate solution with an acid selected from the group consisting of organic, mineral and oxy acids to a final pH of about  
10 5.0 to 10.5 to provide a reactive silica;

(d) aging reactive silica from step (b) at temperatures greater than -20°C;

(e) recovering a solid product from the aqueous solution; and

15 (f) removing the surfactant by removal of the solution to provide the molecular sieve silica, wherein the silica possesses framework-confined mesopores with pore diameters ranging from 1.0 to 12.0 nm, the framework-confined channel structure comprises the hybrid  
20 of a wormhole and lamellar or wormhole framework morphology, has one resolved powder X-ray reflection corresponding to a pore-pore correlation spacing of 1.5 to 15.0 nm, inorganic oxide wall thickness of greater than 0.5 nm, specific surface areas of 400 to 1400 m<sup>2</sup>/g  
25 and framework pore volumes of 0.2 to 3.0 cc/g N<sub>2</sub>.

-36-(Original)

A composition which is a hybrid molecular sieve silica prepared by a process which comprises:

(a) acidifying an aqueous solution of an amine surfactant containing an alkyl chain with 6 to 36 carbon atoms as the organic structure director with an acid selected from the group consisting of organic, mineral and oxy acids;

(b) preparing a reactive silica species by addition of a soluble silicate to the acidified amine surfactant reaching a pH of less than 4;

(c) titrating the reactive silica with a base to a final pH of about 5.0 to 10.5;

(d) aging reactive silica from step (b) at temperatures greater than -20°C;

(e) recovering a solid product from the aqueous solution; and

(f) removing the surfactant from the solid product to provide the molecular sieve silica, wherein the resulting inorganic oxide possesses framework-confined mesopores with pore diameters ranging from 10 to 12.0 nm, the framework-confined channel structure comprises the hybrid of a wormhole and lamellar or hexagonal framework morphology, has at least one resolved powder x-ray reflection corresponding to a pore-pore correlation spacing of 1.5 to 15.0 nm, inorganic oxide

wall thickness of greater than 0.5 nm, specific surface areas of 400 to 1400 m<sup>2</sup>/g and framework pore volumes of 0.2 to 3.0 cc/g N<sub>2</sub>.

5

-37-(Original)

A process for the preparation of a hybrid wormhole and lamellar or hexagonal molecular sieve silica which comprises:

10

(a) reacting in an aqueous solution, an amine surfactant and a reactive silica species of pH between 5.0 and 10.5;

(b) aging the solution to precipitate the silica; and

15

(c) removing the silica from the solution.

-38-(Original)

A process for the preparation of a hybrid molecular sieve silica which comprises:

(a) providing a protonated amine surfactant solution with a pH below 7.0;

5 (b) reacting the protonated amine surfactant solution with a mixture of a base and a soluble silicate solution to produce a reactive silica species at a final pH between about 5.0 and 10.5;

10 (c) aging the reactive silica species in the solution of step (b) at a temperature greater than -20°C to form a precipitated product which is the silica in the solution; and

(d) recovering the precipitated product from the solution.

-39-(Original)

The process of Claim 38 wherein the surfactant is removed from the precipitated product.



-40- (Original)

A process for the preparation of a hybrid molecular sieve silica which comprises:

(a) acidifying surfactant solution of a neutral amine surfactant with an acid thereof to produce a pH below 7.0;

(b) forming a reactive silica species by neutralization of a soluble silicate solution with the surfactant solution of step (a) to provide a final pH of about 5.0 to 10.5;

(c) aging the reactive silica species in the solution of step (b) at a temperature greater than  $-20^{\circ}\text{C}$  to form a precipitated product which is the silica composition in the solution; and

(d) recovering the precipitated product from the solution.

-41- (Original)

The process of Claim 40 wherein soluble silica solution is a sodium silicate with  $\text{SiO}_2/\text{OH}^-$  ratio of between 0.7 and 2.

-42- (Original)

The process of Claim 40 wherein the acid is an organic acid.

-43-(Original)

The process of Claim 42 wherein the acid is selected from the group consisting of acetic, glycolic, formic and citric acid.

-44-(Original)

The process of Claim 40 wherein the surfactant is removed by calcination, solvent extraction or acid washing.

-45-(Original)

The process of Claim 40 with the additional step (d) of removing the surfactant and by calcination of the precipitated product in air for not less than 30 minutes.

-46-(Original)

A process for the preparation of a hybrid molecular sieve silica which comprises:

(a) providing an aqueous solution of a water soluble silicate at a pH greater than 9;

5 (b) combining the aqueous solution with a neutral amine surfactant and an acid to produce a resulting mixture wherein the pH of the mixture is between about 5.0 and 10.5;

10 (c) aging the resulting mixture at a temperature between -20° and 100°C until the hybrid molecular sieve silica is formed; and

(d) removing at least the aqueous solution to produce the hybrid molecular sieve silica.

-47-(Original)

A process for the preparation of a hybrid molecular sieve aluminosilicate which comprises:

5 (a) providing an aqueous solution of a water soluble aluminate and silicate in a molar ratio of aluminate to silicate of between about 0.01 and 1.0 at a pH greater than 9;

10 (b) combining the aqueous solution with a neutral amine surfactant and an acid in aqueous solution to produce a resulting mixture wherein the pH of the mixture to be between about 5.0 and 10.5;

(c) aging the resulting mixture at a temperature between -20° and 100°C until the hybrid molecular sieve aluminosilicate is formed; and

15 (d) removing at least the aqueous solution to produce the hybrid molecular sieve aluminosilicate.

-48-(Original)

A process for the preparation of a hybrid molecular sieve aluminosilicate which comprises:

(a) providing an aqueous solution of a water soluble silicate at a pH greater than 9;

5 (b) combining the aqueous solution with a neutral amine surfactant, an aluminum salt and an acid in aqueous solution to produce a resulting mixture wherein the aluminum to silicon molar ratio is between 0.01 and 1.0 and the pH of the mixture to be between about 5.0 and  
10 10.5;

(c) aging the resulting mixture at a temperature between -20° and 100°C until the hybrid molecular sieve aluminosilicate is formed; and

(d) removing at least the aqueous solution to  
15 produce the hybrid molecular sieve aluminosilicate.

-49-(Original)

The process of Claim 48 wherein in step (d) the surfactant and water are removed from the aluminosilicate so that aluminosilicate is dry.

-50-(Original)

The process of Claim 48 wherein the aluminosilicate is calcined.

-51-(Original)

The process of Claim 48 wherein the aluminum salt is selected from the group consisting of aluminum nitrate, aluminum chloride, aluminum sulfate and a cationic aluminum oligomer.

-52-(Original)

The composition of Claim 1 derived from a mixture of a neutral amine, basic silicate and acid in an aqueous solution to produce a pH between about 5 to 10.5.

-53-(Original)

The composition of Claim 1 derived from a mixture of a protonated amine and a basic silicate in an aqueous solution to produce a pH between about 5.0 and 10.5.